

PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2003-007333

(43)Date of publication of application : 10.01.2003

(51)Int.Cl.

H01M 10/40

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(54) NONAQUEOUS ELECTROLYTIC SOLUTION AND NONAQUEOUS ELECTROLYTIC SOLUTION SECONDARY BATTERY USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a nonaqueous electrolytic solution secondary battery and a nonaqueous electrolytic solution for use in the same, wherein the secondary battery has a high charging/discharging efficiency, superior capacity retaining characteristics, excellent battery characteristics and safety over a wide range of temperatures, and a high energy density.

SOLUTION: A nonaqueous solution containing lactone compounds and nitrogen-containing heterocycle compounds is provided, wherein the lactone compound is selected from γ -butyrolactone, γ -valerolactone, δ -valerolactone, γ -caprolactone, δ -caprolactone, and ϵ -caprolactone, and the nitrogen-containing heterocycle compound is represented by a general formula (1) $A(N-R)_n$ (A stands for nitrogen-containing heterocycle containing carbonyl group, R for alkyl group, alkenyl group, or (hetero-)aryl group, and n for a natural number. If n represents 2 or a larger number, however, R may represent a compound different from each compound described above.) Lithium salt, as an electrolyte, is dissolved in the nonaqueous solution to make the nonaqueous electrolytic solution, which is used for the nonaqueous electrolytic solution secondary battery.

LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of
rejection]

[Kind of final disposal of application other

than the examiner's decision of rejection or
application converted registration]

[Date of final disposal for application]

[Patent number]

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examiner's decision of rejection]

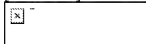
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CLAIMS

[Claim(s)]

[Claim 1] A lactone compound and a general formula (1)

[Formula 1]



(A expresses among a formula the nitrogen-containing heterocycle which has a carbonyl group, R expresses an alkyl group, an alkenyl radical, or (hetero) an aryl group, and n expresses the natural number.) However, when n is two or more, R may differ, respectively. Nonaqueous electrolyte characterized by dissolving lithium salt in the non-aqueous solvent containing the nitrogen-containing heterocycle compound expressed.

[Claim 2] The nonaqueous electrolyte according to claim 1 to which a non-aqueous solvent is characterized by including 10 - 99.8 % of the weight, and a nitrogen-containing heterocycle compound for a lactone compound 0.1 to 10% of the weight.

[Claim 3] The nonaqueous electrolyte according to claim 1 or 2 characterized by choosing a lactone compound from the group which consists of gamma-butyrolactone, gamma-valerolactone, delta-valerolactone, gamma-caprolactone, a delta-caprolactone, and an epsilon-caprolactone.

[Claim 4] The nonaqueous electrolyte according to claim 1 to 3 characterized by choosing a nitrogen-containing heterocycle compound from the group which consists of a 1-alkyl pyrrolidone compound, 1-alkyl piperidone compound, 3-alkyl oxazolidone compound, 1, 3-dialkyl imidazolidinone compound, 1, and 3-dialkyl hexahydro pyrimidinone compound and an N-alkyl OKISAJINON compound.

[Claim 5] A nitrogen-containing heterocycle compound A 1-methyl-2-pyrrolidone, a 1-vinyl-2-pyrrolidone, 3-methyl-2-oxazolidone, 3-vinyl-2-oxazolidone, 1,3-dimethyl-2-imidazolidinone, 1, 3-divinyl-2-imidazolidinone, 1, 3-dimethyl hexahydro pyrimidine-2-ON, 1, 3-divinyl hexahydro pyrimidine-2-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1 and 3-oxazine-2-ON and 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - It is what was chosen from the group which consists of 1 and 3-oxazine-2-ON. The nonaqueous electrolyte according to claim 1 to 3 characterized by a certain thing.

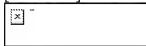
[Claim 6] lithium salt -- LiPF₆, LiBF₄, and LiCF₃ -- the nonaqueous electrolyte according to claim 1 to 5 characterized by being chosen out of the group which consists of SO₃, LiN (CF₃SO₂)₂, LiN (C₂F₅SO₂)₂ and LiN (CF₃SO₂) (C₄F₉SO₂), LiPF₃(C₂F₅)₃, and LiB (CF₃COO)₄.

[Claim 7] The nonaqueous electrolyte according to claim 1 to 6 characterized by containing the coat generation agent chosen from the group which a non-aqueous solvent becomes from vinylene carbonate, vinyl ethylene carbonate, phenylethylene carbonate, an ethylene aprotic solvent, a propane sultone, a succinic anhydride, an anhydrous malonic acid, a maleic anhydride, and phthalic anhydride.

[Claim 8] The nonaqueous electrolyte according to claim 7 characterized by a non-aqueous solvent containing 0.1 - 5 % of the weight for a coat generation agent.

[Claim 9] gamma-butyrolactone -- 10 - 99.9 % of the weight; -- ethylene carbonate -- Propylene carbonate, butylene carbonate, dimethyl carbonate, Diethyl carbonate, G n-propyl carbonate, diisopropyl carbonate, n-propyl isopropyl carbonate, di-n-butyl carbonate, Diisobutyl carbonate, G t-butyl carbonate, n-butyl isobutyl carbonate, n-butyl-t-butyl carbonate, isobutyl-t-butyl carbonate, Ethyl methyl carbonate, methyl-n-propyl carbonate, n-butyl methyl carbonate, Isobutyl methyl carbonate, t-butyl methyl carbonate, ethyl-n-propyl carbonate, n-butyl ethyl carbonate, isobutyl ethyl carbonate, t-butyl ethyl carbonate, n-butyl-n-propyl carbonate, isobutyl-n-propyl carbonate, t-butyl-n-propyl carbonate, n-butyl isopropyl carbonate, Isobutyl isopropyl carbonate, t-butyl isopropyl carbonate, Methyl acetate, ethyl acetate, n propyl acetate, acetic-acid-isopropyl, N-butyl acetate, isobutyl acetate, acetic-acid-t-butyl, methyl propionate, Ethyl propionate, propionic-acid-n-propyl, propionic-acid-isopropyl, Propionic-acid-n-butyl, propionic-acid isobutyl, propionic-acid-t-butyl, It is a 0-89.9 % of the weight; general formula (1) about what was chosen from the group which consists of dimethoxymethane, 1, 2-dimethoxyethane, 1, 2-diethoxy methane, diethoxy ethane, and ethoxy methoxy methane.

[Formula 2]



(A expresses among a formula the nitrogen-containing heterocycle which has a carbonyl group, R expresses an alkyl group, an alkenyl radical, or (hetero) an aryl group, and n expresses the natural number.) However, when n is two or more, R may differ, respectively. The nitrogen-containing heterocycle compound expressed 0.1 - 10 % of the weight, Vinylene carbonate, vinyl ethylene carbonate, phenylethylene carbonate, An ethylene aprotic solvent, a propane sultone, a succinic anhydride, an anhydrous malonic acid, To the non-aqueous solvent contained zero to 5% of the weight, the compound chosen from the group which consists of a maleic anhydride and phthalic anhydride LiPF₆, LiBF₄, and LiCF₃ -- SO₃ and LiN (CF₃SO₂)₂ -- The nonaqueous electrolyte characterized by dissolving the lithium salt chosen from LiN (C₂F₅SO₂)₂, LiN (CF₃SO₂) (C₄F₉SO₂), LiPF₃(C₂F₅)₃, and the group that consists of LiB (CF₃COO)₄. [Claim 10] The nonaqueous electrolyte rechargeable battery characterized by becoming a positive electrode containing occlusion and the ingredient which can be emitted from the electrolytic solution according to claim 1 to 9 about the negative electrode which contains occlusion and the ingredient which can be emitted for a metal lithium, a lithium alloy, or a lithium, and a lithium.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the nonaqueous electrolyte rechargeable battery which used a nonaqueous electrolyte and this. In detail, charge-and-discharge effectiveness is high and it is related with the nonaqueous electrolyte which gives the nonaqueous electrolyte rechargeable battery of a high energy consistency and it which were excellent in the capacity maintenance property, and excellent in the cell property and safety in a large temperature requirement.

[0002]

[Description of the Prior Art] development of the lithium secondary battery which has a high energy density with lightweight-izing of an electric product in recent years, and a miniaturization -- before -- furthermore, it is wished and the improvement of a cell property is also demanded with expansion of Field of application of a lithium secondary battery. In the lithium secondary battery, disassembly of the solvent of the electrolytic solution takes place in an electrode surface, and this has become charge-and-discharge effectiveness and the cause of a fall of capacity maintenance. For example, if the electrolytic solution which used propylene carbonate as the main solvent is used, the decomposition reaction of a solvent occurs on a negative-electrode front face, and it is known for the nonaqueous electrolyte rechargeable battery which mixed the various electrode material of a graphite system with independence, mixed the lithium with occlusion and the negative-electrode material which can be emitted, and was used as the negative electrode that the occlusion of the smooth lithium to a graphite and emission will become impossible.

[0003] On the other hand, although there is little decomposition compared with propylene carbonate when ethylene carbonate is used as an electrolytic-solution solvent, at a room temperature, ethylene carbonate is a solid-state, and since viscosity is high, it has the fault of being hard to deal with it. Moreover, although gamma-butyrolactone has the outstanding property which a non-aqueous solvent for nonaqueous electrolytes called hypoviscosity and a high dielectric constant is expected, it has the fault that electrochemical decomposition arises on a positive/negative two-electrodes front face. It is indicated by JP, 11-31525, A the electrolytic solution which contains the ethylene carbonate of 15 - 35 capacity % extent as a subsolvent, using gamma-butyrolactone as a main solvent that disassembly of the solvent on an electrode surface can be suppressed.

[0004]

[Problem(s) to be Solved by the Invention] Since the electrolytic solution which used gamma-butyrolactone and ethylene carbonate together was inferior to electrochemical oxidation resistance and reducibility-proof as compared with the electrolytic solution which used together ethylene carbonate and hypoviscosity solvents other than gamma-butyrolactone, there is a problem in the capacity maintenance factor of the cell at the time of an elevated temperature etc., and the further amelioration was desired.

[0005] Moreover, since the presentation of the electrolytic solution affects the charge-and-discharge property and capacity maintenance property of a lithium secondary battery, making the electrolytic solution contain various compounds and improving an above-mentioned property is examined (refer to JP,3-57169,A, 8-321312, 10-14529, 11-273728, and JP,2000-285962,A each official report). However, about the electrolytic-solution solvent which contains a lactone compound in a solvent, the effective compound is not yet known.

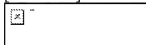
[0006] This invention tends to offer the nonaqueous electrolyte which has the cell properties of having excelled in the large temperature requirement, such as a charge-and-discharge property and a capacity maintenance property, and gives a nonaqueous electrolyte rechargeable battery with high safety.

[0007]

[Means for Solving the Problem] this invention person etc. is a lactone compound and a general formula (1), as a result of repeating various examination, in order to solve the above-mentioned technical problem.

[0008]

[Formula 3]



[0009] (A expresses among a formula the nitrogen-containing heterocycle which has a carbonyl group, R expresses an alkyl group, an alkenyl radical, or (hetero) an aryl group, and n expresses the natural number.) However, when n is two or more, R may differ, respectively. To the non-aqueous solvent containing the nitrogen-containing heterocycle compound expressed The lithium secondary battery using the nonaqueous electrolyte in which lithium salt was dissolved Since a coat with sufficient stability generates from the time of early charge efficiently with lithium ion permeability to an electrode surface and disassembly of the electrolytic solution is controlled In the large temperature requirement, it finds out excelling in cell properties, such as charge-and-discharge effectiveness and a capacity maintenance property, and having high safety, and came to complete this invention.

[0010]

[Embodiment of the Invention] In this invention, what contains a lactone compound as a solvent of the electrolytic solution is used. As a lactone compound, 5 thru/or 7 membered-ring lactone, such as gamma-butyrolactone, gamma-valerolactone, delta-valerolactone, gamma-caprolactone, delta-caprolactone, and epsilon-caprolactone, can usually be used. Especially, gamma-butyrolactone is desirable. A lactone compound can also use some together.

[0011] A lactone compound is used so that 10 - 99.9% of the weight of a non-aqueous solvent may be occupied. It is desirable to use 12 to 99% of the weight, so that 15 - 98 % of the weight may be occupied especially. To a lactone compound, other non-aqueous solvents regularly used by the nonaqueous electrolyte can also be used together. As such a non-aqueous solvent, annular carbonate and chain-like carbonate, carboxylate, the chain-like ether, etc. are mentioned.

[0012] As annular carbonate, ethylene carbonate, propylene carbonate, butylene carbonate, vinylene carbonate, vinyl ethylene carbonate, etc. are mentioned. As chain-

like carbonate, dimethyl carbonate, diethyl carbonate, G n-propyl carbonate, diisopropyl carbonate, n-propyl isopropyl carbonate, Di-n-butyl carbonate, diisobutyl carbonate, G t-butyl carbonate, n-butyl isobutyl carbonate, n-butyl-t-butyl carbonate, Isobutyl-t-butyl carbonate, ethyl methyl carbonate, Methyl-n-propyl carbonate, n-butyl methyl carbonate, Isobutyl methyl carbonate, t-butyl methyl carbonate, ethyl-n-propyl carbonate, n-butyl ethyl carbonate, isobutyl ethyl carbonate, t-butyl ethyl carbonate, n-butyl-n-propyl carbonate, isobutyl-n-propyl carbonate, t-butyl-n-propyl carbonate, n-butyl isopropyl carbonate, Isobutyl isopropyl carbonate, t-butyl isopropyl carbonate, etc. are used, and dimethyl carbonate, diethyl carbonate, and ethyl methyl carbonate are desirable especially.

[0013] As carboxylate, methyl acetate, ethyl acetate, n propyl acetate, acetic-acid-isopropyl, n-butyl acetate, isobutyl acetate, acetic-acid-t-butyl, methyl propionate, ethyl propionate, propionic-acid-n-propyl, propionic-acid-isopropyl, propionic-acid-n-butyl, propionic-acid isobutyl, propionic-acid-t-butyl, etc. are used, and ethyl acetate, methyl propionate, and ethyl propionate are desirable especially.

[0014] As the chain-like ether, dimethoxymethane, 1, 2-dimethoxyethane, diethoxy methane, 1, 2-diethoxy ethane, ethoxy methoxy methane, ethoxy methoxyethane, etc. are used, and 1, 2-dimethoxyethane, 1, and 2-diethoxy ethane is desirable especially. These solvents are used so that 0 - 89.9% of the weight of a non-aqueous solvent may be occupied.

[0015] The nitrogen-containing heterocycle compound expressed with a general formula (1) to a non-aqueous solvent is made to contain in this invention. In the compound of a general formula (1), the nitrogen-containing heterocycle which has the carbonyl group expressed with A may also condense the monocycle with the benzene ring etc. The endocyclic numbers of carbonyl groups are 1-4. As an example of nitrogen-containing heterocycle, a pyrrolidine, imidazolidine, oxazolidine, thiazolidine, a piperidine, a pyrimidine, oxazine, indoline (ISO), benzimidazole, benzoxazole, benzothiazole, a quinoline, quinazoline, benzoxazine, etc. are mentioned.

[0016] In the compound of a general formula (1), when R expresses an alkyl group, the alkyl group is an alkyl group of the carbon numbers 1-4 which may have the substituent, and is a methyl group or an ethyl group preferably. When R expresses an alkenyl radical, the alkenyl radical is an alkenyl radical of the carbon numbers 2-4 which may have the substituent, and is a vinyl group preferably.

[0017] When R expresses an aryl group (hetero), the (hetero) aryl group is the phenyl group or pyridyl radical which may have the substituent. The following compounds are mentioned as an example of a nitrogen-containing heterocycle compound expressed with a general formula (1).

1) A pyrrolidine compound 1-methyl-2-pyrrolidone, a 1-ethyl-2-pyrrolidone, A 1-vinyl-2-pyrrolidone, 1, a 5-dimethyl-2-pyrrolidone, A 1-isopropyl-2-pyrrolidone, a 1-n-butyl-2-pyrrolidone, 1-alkyl (or alkenyl) pyrrolidone compound; 1-phenyl-2-pyrrolidones, such as a 1-methyl-3-pyrrolidone, a 1-ethyl-3-pyrrolidone, and a 1-vinyl-3-pyrrolidone, 1-aryl pyrrolidone compounds, such as a 1-phenyl-3-pyrrolidone; N-methyl succinimide, N-alkenyl succinimide compound; N-phenyl succinimides, such as N-alkyl succinimide compound; N-vinyl succinimides, such as N-ethyl succinimide, N-cyclohexyl succinimide, and N-isobutyl succinimide, N-(hetero) aryl succinimide compounds, such as N-(p-tolyl) succinimide and N-(3-pyridyl) succinimide.

- 2) ISO indoline compound 1-methyl-2-in DORINON, 1-ethyl-2-in DORINON, 1-vinyl-2-in DORINON, 1-methyl-3-in DORINON, 1-aryl in DORINON compound; N-methyl phthalimides [, such as 1-alkyl (or alkenyl) in DORINON compound; 1-phenyl-2-in DORINON], such as 1-ethyl-3-in DORINON and 1-vinyl-3-in DORINON, N-alkyl phthalimide compound [, such as N-ethyl phthalimide,]; -- N-alkenyl phthalimide compound [, such as N-aryl phthalimide,]; -- N-aryl phthalimide compounds, such as N-phenyl phthalimide.
- 3) The imidazolidine compound 1, 3-dimethyl imidazolidine -2, 5-dione, The 1-ethyl-3-methylimidazo lysine -2, 5-dione, 1, 3-diethyl imidazolidine -2, 5-dione, 1-alkyl-3-aryl imidazolidine -2, such as 1, such as 1, 3-divinyl imidazolidine -2, and 5-ON, 3-dialkyl (or alkenyl) imidazolidine -2, 5-dione compound; 1-methyl-3-phenyl imidazolidine -2, and 5-dione, 5-dione compound; -- 1, such as 1, 3-diphenyl imidazolidine -2, and 5-dione, 3-diaryl imidazolidine -2, and 5-dione compound; 1,3-dimethyl-2-imidazolidinone -- 1-ethyl-3-methyl-2-imidazolidinone, 1, 3-diethyl-2-imidazolidinone, 1, 3-divinyl-2-imidazolidinone, 1, 3-dimethyl-4-imidazolidinone, 1-ethyl-3-methyl-4-imidazolidinone, 3-ethyl-1-methyl-4-imidazolidinone, 1, such as 1, 3-diethyl-4-imidazolidinone, 1, and 3-divinyl-4-imidazolidinone, 3-dialkyl (or alkenyl) imidazolidinone compound; 1-methyl-3-phenyl-2-imidazolidinone, 1(3)-alkyl-3(1)-aryl imidazolidinone compounds, such as 1-methyl-3-phenyl-4-imidazolidinone and 3-methyl-1-phenyl-4-imidazolidinone; 1, 3-diphenyl-2-imidazolidinone, 1, such as 1 and 3-diphenyl-4-imidazolidinone, a 3-diaryl imidazolidinone compound.
- 4) The benzimidazole compound 1, the 3-dimethyl -2, 3-dihydrobenzimidazole-2-ON, The 1-ethyl-3-methyl -2, 3-dihydrobenzimidazole-2-ON, 1, the 3-diethyl -2, 3-dihydrobenzimidazole-2-ON, 1, the 3-divinyl -2, 1, such as 3-dihydrobenzimidazole-2-ON, the 3-dialkyl (or alkenyl) -2, 3-dihydrobenzimidazole - 1-alkyl-3-aryls -2, such as the 2-ON compound; 1-methyl-3-phenyl -2 and 3-dihydrobenzimidazole-2-ON, 3-dihydrobenzimidazole-2-ON compound; 1, such as 1, 3-diphenyl -2, and 3-dihydrobenzimidazole-2-ON, 3-diaryl - 2 and 3-dihydrobenzimidazole-2-ON compound.
- 5) Oxazolidine compound 3-methyl-2-oxazolidone, 3-ethyl-2-oxazolidone, 3-methyl-4-oxazolidone, 3-ethyl-4-oxazolidone, 3-alkyl oxazolidone compound; 3-vinyl-2-oxazolidone, such as 3-methyl-5-oxazolidone and 3-ethyl-5-oxazolidone, 3-alkenyl oxazolidone compound; 3-phenyl-2-oxazolidone, such as 3-vinyl-4-oxazolidone and 3-vinyl-5-oxazolidone, 3-aryl oxazolidone compound; 3-methyl oxazolidine -2, such as 3-phenyl-4-oxazolidone and 3-phenyl-5-oxazolidone, 4-dione, 3-ethyl oxazolidine -2, 4-dione, 3-vinyl oxazolidine -2, 4-dione, 3-methyl oxazolidine -2, 5-dione, 3-ethyl oxazolidine -2, 5-dione, 3-aryl oxazolidinedione compounds, such as 3-alkyl (or alkenyl) oxazolidinedione compound; 3-phenyl oxazolidine -2, such as 3-vinyl oxazolidine -2 and 5-dione, 4-dione, 3-phenyl oxazolidine -2, and 5-dione.
- 6) The benzooxazole compound 3-methyl -2, 3-dihydrobenzooxazole-2-ON, 3-ethyl -2, 3-dihydrobenzooxazole-2-ON, The 3-aryls -2, such as the 3-alkyls (or alkenyl) -2, such as 3-vinyl -2 and 3-dihydrobenzooxazole-2-ON, the 3-dihydrobenzooxazole compound; 3-phenyl -2, and 3-dihydrobenzooxazole-2-ON, 3-dihydrobenzooxazole-2-ON compound.
- 7) Thiazolidine compound 3-methyl-2-thiazolidone, 3-ethyl-2-thiazolidone, 3-vinyl-2-thiazolidone, 3-methyl-4-thiazolidone, 3-ethyl-4-thiazolidone, 3-vinyl-4-thiazolidone, 3-methyl-5-thiazolidone, 3-ethyl-5-thiazolidone, 3-alkyl (or alkenyl) thiazolidone

compounds, such as 3-vinyl-5-thiazolidone; 3-phenyl-2-thiazolidone, 3-aryl thiazolidone compound; 3-methyl thiazolidine -2, such as 3-phenyl-4-thiazolidone and 3-phenyl-5-thiazolidone, 4-dione, 3-ethyl thiazolidine -2, 4-dione, 3-vinyl thiazolidine -2, 4-dione, 3-methyl thiazolidine -2, 5-dione, 3-ethyl thiazolidine -2, 5-dione, 3-aryl thiazolidinedione compounds, such as 3-alkyl (or alkenyl) thiazolidinedione compound; 3-phenyl thiazolidine -2, such as 3-vinyl thiazolidine -2 and 5-dione, 4-dione, 3-phenyl thiazolidine -2, and 5-dione.

8) The 3-aryls -2, such as the 3-alkyls (or alkenyl) -2, such as the benzothiazole compound 3-methyl -2, 3-dihydrobenzothiazole-2-ON, 3-vinyl -2, and 3-dihydrobenzothiazole-2-ON, the 3-dihydrobenzothiazole-2-ON compound; 3-phenyl -2, and 3-dihydrobenzothiazole-2-ON, a 3-dihydrobenzothiazole-2-ON compound.

9) Piperidine compound 1-methyl-2-piperidone, 1-ethyl-2-piperidone, 1-vinyl-2-piperidone, 1-methyl-3-piperidone, 1-ethyl-3-piperidone, 1-vinyl-3-piperidone, 1-methyl-4-piperidone, 1-ethyl-4-piperidone, 1-vinyl-4-piperidone, 1-morpholino methyl-2-piperidone, 1-(permutation) alkyl (or alkenyl) piperidone compounds, such as 1-piperidinomethyl-2-piperidone; 1-aryl piperidone compounds, such as 1-phenyl-2-piperidone, 1-phenyl-3-piperidone, and 1-phenyl-4-piperidone.

10) The quinoline compound 3, 4-dihydro-1-methyl-2-quinolone, 3, such as 3 and 4-dihydro-1-methyl-3-quinolone, 3, and 4-dihydro-1-methyl-quinoline-4-ON, 4-dihydro-1-alkyl quinolone compound; 3, 4-dihydro-1-phenyl-2-quinolone, 3, such as 3 and 4-dihydro-1-phenyl-3-quinolone, 3, and 4-dihydro-1-phenyl-4-quinolone, a 4-dihydro-1-aryl quinolone compound.

11) The pyrimidine compound 1, 3-dimethyl hexahydro pyrimidine-2-ON, 1, 3-diethyl hexahydro pyrimidine-2-ON, 1, 3-divinyl hexahydro pyrimidine-2-ON, 1, 3-dimethyl hexahydro pyrimidine-4-ON, 1-ethyl-3-methyl hexahydro pyrimidine-4-ON, 3-ethyl-1-methyl hexahydro pyrimidine-4-ON, 1, 3-diethyl hexahydro pyrimidine-4-ON, 1, 3-divinyl hexahydro pyrimidine-4-ON, 1, 3-dimethyl hexahydro pyrimidine-5-ON, 1-ethyl-3-methyl hexahydro pyrimidine-5-ON, 1, 3-diethyl hexahydro pyrimidine-5-ON, 1, such as 1 and 3-divinyl hexahydro pyrimidine-5-ON, 3-dialkyl (or alkenyl) hexahydro pyrimidinone compound; 1-methyl-3-phenyl hexahydro pyrimidine-2-ON, 1-methyl-3-phenyl hexahydro pyrimidine-4-ON, 3-methyl-1-phenyl hexahydro pyrimidine-4-ON, 1-alkyl-3-aryl hexahydro pyrimidinone compounds, such as 1-methyl-3-phenyl hexahydro pyrimidine-5-ON; 1, 3-diphenyl hexahydro pyrimidine-2-ON, 1, such as 1, 3-diphenyl hexahydro pyrimidine-4-ON, 1, and 3-diphenyl hexahydro pyrimidine-5-ON, a 3-diaryl hexahydro pyrimidinone compound; 1, 3-dimethyl hexahydro pyrimidine -2, 4-dione, 1, 3-diethyl hexahydro pyrimidine -2, 4-dione, the 1-ethyl-3-methyl-hexahydro pyrimidine -2, 4-dione, The 3-ethyl-1-methyl-hexahydro pyrimidine -2, 4-dione, 1, 3-divinyl hexahydro pyrimidine -2, 4-dione, 1, 3-dimethyl hexahydro pyrimidine -2, 5-dione, The 1-ethyl-3-methyl-hexahydro pyrimidine -2, 5-dione, 1, 3-diethyl HEKISADORO pyrimidine -2, 5-dione, 1, such as 1, 3-divinyl hexahydro pyrimidine -2, and 5-dione, the 3-dialkyl (or alkenyl) hexahydro pyrimidine dione compound; 1-methyl-3-phenyl hexahydro pyrimidine -2, 4-dione, The 3-methyl-1-phenyl hexahydro pyrimidine -2, 4-dione, 1(3)-alkyl-3(1)-aryl hexahydro pyrimidine dione compounds, such as the 1-methyl-3-phenyl hexahydro pyrimidine -2 and 5-dione; 1, 3-diphenyl hexahydro pyrimidine -2, 4-dione, 1, such as 1, 3-diphenyl hexahydro pyrimidine -2, and 5-dione, a 3-diaryl hexahydro pyrimidine dione compound; 1, 3-dimethyl hexahydro pyrimidine -2,

4, 5-trione, 1, 3-diethyl hexahydro pyrimidine - 2, 4, 5-trione, 1-ethyl-3-methyl hexahydro pyrimidine - 2, 4, 5-trione, 3-ethyl-1-methyl hexahydro pyrimidine - 2, 4, 5-trione, 1, 3-divinyl hexahydro pyrimidine - 2, 4, 5-trione, 1, 3-dimethyl hexahydro pyrimidine - 2, 4, 6-trione, 1, 3-diethyl hexahydro pyrimidine - 2, 4, 6-trione, 1-ethyl-3-methyl hexahydro pyrimidine - 2, 4, 6-trione, 3-ethyl-1-methyl hexahydro pyrimidine - 2, 4, 6-trione, 1, 3-divinyl hexahydro pyrimidine - 1, such as 2, 4, and 6-trione, 3-dialkyl (or alkenyl) hexahydro pyrimidine trione compound; 1-methyl-3-phenyl hexahydro pyrimidine - 2, 4, 5-trione, 3-methyl-1-phenyl hexahydro pyrimidine - 2, 4, 5-trione, 1, 3-dimethyl hexahydro pyrimidine TETORAON, 1, 3-diethyl hexahydro pyrimidine TETORAON, 1-ethyl-3-methyl hexahydro pyrimidine TETORAON, The 1(3)-alkyls -3, such as 1, such as 1 and 3-divinyl hexahydro pyrimidine TETORAON, and 3-dialkyl (or alkenyl) hexahydro pyrimidine tetrapod-on compound; 1-methyl-3-phenyl hexahydro pyrimidine TETORAON (1) - aryl hexahydro pyrimidine TETORAON compound; -- a 1, such as 1 and 3-diphenyl hexahydro pyrimidine TETORAON, and 3-diaryl hexahydro pyrimidine TETORAON compound.

12) The quinazoline compound 1, 3-dimethyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1, 3-diethyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1-ethyl-3-methyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 3-ethyl-1-methyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1, 3-divinyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1, 3-dimethyl - 1, 2, 3, 4-tetrahydro quinazoline-4-ON, 1, 3-diethyl - 1, 2, 3, 4-tetrahydro quinazoline-4-ON, 1-ethyl-3-methyl - 1, 2, 3, 4-tetrahydro quinazoline-4-ON, 3-ethyl-1-methyl - 1, 2, 3, 4-tetrahydro quinazoline-4-ON, 1, 3-divinyl [- 1, 2, 3, 4-tetrahydro quinazoline-2-ON,] - 1, such as 1, 2, 3, and 4-tetrahydro quinazoline-4-ON, 3-dialkyl (or alkenyl) - 1, 2, 3, 4 - Tetrahydro chinae-cortex ZORINON compound; 1-methyl-3-phenyl 3-methyl-1-phenyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1-methyl-3-phenyl - 1, 2, 3, 4-tetrahydro quinazoline-4-ON, The 1(3)-alkyl-3(1)-aryls -1, such as 3-methyl-1-phenyl -1, 2 and 3, and 4-tetrahydro quinazoline-4-ON, 2 and 3, a 4-tetrahydro chinae-cortex ZORINON compound; 1, 3-diphenyl - 1, 2, 3, 4-tetrahydro quinazoline-2-ON, 1, such as 1, 3-diphenyl -1, 2 and 3, and 4-tetrahydro quinazoline-4-ON, 3-diaryl-1, 2 and 3, a 4-tetrahydro chinae-cortex ZORINON compound; 1, 3-dimethyl - 1, 2, 3, 4-tetrahydro quinazoline - 2, 4-dione, 1, 3-diethyl - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione, 1-ethyl-3-methyl - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione, 3-ethyl-1-methyl - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione, 1, 3-divinyl - 1, such as 1, 2, 3, 4-tetrahydro quinazoline -2, and 4-dione, 3-dialkyl (or alkenyl) - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione compound; 1-methyl-3-phenyl - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione, The 1(3)-alkyl-3(1)-aryls -1, such as the 3-methyl-1-phenyl -1, 2 and 3, 4-tetrahydro quinazoline -2, and 4-dione, 2 and 3, 4-tetrahydro quinazoline -2, 4-dione compound; 1, 3-diphenyl - 1, 2, 3, 1, such as 4-tetrahydro quinazoline -2 and 4-dione, 3-diaryl - 1, 2, 3, 4-tetrahydro quinazoline -2, 4-dione compound.

13) Oxazine compound 2-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-3-ON, 2-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-3-ON, 2-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-3-ON, 2-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-4-

ON, 2-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-4-ON, 2-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-4-ON, 2-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-5-ON, 2-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-5-ON, 2-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-5-ON, 2-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-6-ON, 2-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-6-ON, 2-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-6-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-2-ON, 3-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-2-ON, 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-2-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-4-ON, 3-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-4-ON, 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-4-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-5-ON, 3-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-5-ON, 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-5-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-6-ON, 3-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-6-ON, 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-6-ON, 4-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-2-ON, 4-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-2-ON, 4-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-2-ON, 4-methyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-3-ON, 4-ethyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-3-ON, 4-vinyl-2H-3, 4 and 5, 6-tetrahydro - N-alkyl OKISAJINON compound; 2-phenyl-2H-3, such as 1 and 4-oxazine-3-ON, 4 and 5, 6-tetrahydro - 1, 2-oxazine-3-ON, 2-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-4-ON, 2-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-5-ON, 2-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 2-oxazine-6-ON, 3-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-2-ON, 3-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-4-ON, 3-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-5-ON, 3-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 3-oxazine-6-ON, 4-phenyl-2H-3, 4 and 5, 6-tetrahydro - 1, 4-oxazine-2-ON, 4-phenyl-2H-3, 4 and 5, 6-tetrahydro - N-aryl OKISAJINON compounds, such as 1 and 4-oxazine-3-ON.

14) The benzoxazine compound 4-methyl -2, 3-dihydro-4H-1, 4-benzoxazine-2-ON, 4-ethyl -2, 3-dihydro-4H-1, 4-benzoxazine-2-ON, 4-vinyl -2, 3-dihydro-4H-1, 4-benzoxazine-2-ON, The 4-methyl -2, 3-dihydro-4H-1, 4-benzoxazine-3-ON, 4-ethyl -2, 3-dihydro-4H-1, 4-benzoxazine-3-ON, The 4-alkyls (or alkenyl) -1, such as 4-vinyl -2, 3-dihydro-4H-1, and 4-benzoxazine-3-ON, the 4-benzoxazine compound; 4-phenyl -2, 3-dihydro-4H-1, 4-benzoxazine-2-ON, The 4-aryls -1, such as 4-phenyl -2, 3-dihydro-4H-1, and 4-benzoxazine-3-ON, 4-benzoxazine compound.

[0018] In such nitrogen-containing heterocycles, a 1-methyl-2-pyrrolidone, A 1-vinyl-2-pyrrolidone, 3-methyl-2-oxazolidone, 3-vinyl-2-oxazolidone, 1,3-dimethyl-2-imidazolidinone, 1, 3-divinyl-2-imidazolidinone, 1, 3-dimethyl hexahydro pyrimidine-2-ON, 1, 3-divinyl hexahydro pyrimidine-2-ON, 3-methyl-2H-3, 4 and 5, 6-tetrahydro - 1 and 3-oxazine-2-ON and 3-vinyl-2H-3, 4 and 5, 6-tetrahydro - 1 and 3-oxazine-2-ON is desirable.

[0019] Although the nitrogen-containing heterocycle compound expressed with a general formula (1) may be independent or two or more kinds may be used together, the abundance in a non-aqueous solvent is usually used 0.1 to 10% of the weight so that it may become 0.5 - 5 % of the weight preferably. The nitrogen-containing heterocycle compound expressed with a general formula (1) forms a coat with sufficient stability in an electrode surface with lithium ion permeability, and is presumed to be what prevents disassembly of the electrolytic solution. If there is too little abundance in the electrolytic

solution, formation of a coat will become imperfect and expected effectiveness will not fully be discovered. Conversely, if many [too], it will have a bad influence on a cell property.

[0020] The electrolytic solution concerning this invention may contain the still better known coat generation agent, the overcharge inhibitor, the dehydrating agent, the deoxidizer, etc. For example, annular sultones, such as annular ape fight; propane sultones, such as a saturation annular carbonate; ethylene ape fight which has aryl groups, such as saturation annular carbonate; phenylethylene carbonate which has alkenyl radicals, such as partial saturation annular carbonate; vinyl ethylene carbonate, such as vinylene carbonate; coat generation agents, such as annular carboxylic anhydrides, such as a succinic anhydride, an anhydrous malonic acid, a maleic anhydride, and phthalic anhydride, can be made to contain in the electrolytic solution. If such a coat generation agent is contained, charge-and-discharge effectiveness and a capacity maintenance property will become still better. As for a coat generation agent, it is desirable to be added so that it may become 0.1 - 5 % of the weight into a non-aqueous solvent.

[0021] Moreover, for example, JP,8-203560,A, JP,7-302614,A, The benzene derivative indicated by JP,9-50822,A, JP,8-273700,A, and JP,9-17447,A each official report etc.; JP,9-106835,A, JP,9-171840,A, JP,10-321258,A, JP,7-302614,A, JP,7-302614,A, JP,11-162512,A, JP,2939469,B, The biphenyl indicated by JP,2963898,B each official report etc. and its derivative; JP,9-45369,A, The pyrrole derivative indicated by JP,10-321258,A each official report etc.; JP,7-320778,A, The ether system compound indicated by aromatic compound; JP,2983205,B, such as an aniline derivative indicated by JP,7-302614,A each official report etc., etc.; if JP,2001-15158,A is made to contain overcharge inhibitors, such as a publication, now a compound which is, in the electrolytic solution A overcharge condition can be prevented. As for a overcharge inhibitor, it is desirable to make it contain in a non-aqueous solvent, so that it may become 0.1 - 5 % of the weight.

[0022] Lithium salt is used for the solute of the electrolytic solution concerning this invention. The thing of arbitration can be used for lithium salt if it can be used as a solute. As an example, 1 inorganic lithium salt:LiPF₆, LiAsF₆, LiBF₄, LiAlF₄ and LiAlF₆, the inorganic fluoride salt of LiSiF₆ grade, Fault halogen acid salt 2 organic lithium salt of LiClO₄ grade : The organic sulfonate of LiCF₃SO₃ grade, Perfluoroalkyl sulfonic-acid imide salts, such as LiN(CF₃SO₂)₂, and LiN(C₂F₅SO₂)₂, LiN(CF₃SO₂)(C₄F₉SO₂), The perfluoroalkyl sulfonic-acid methide salt of LiC(CF₃SO₂)₃ grade, The salt which permuted some fluorines of inorganic fluoride salts, such as LiPF₃(C₂F₅)₃, LiBF₂(CF₃)₂, and LiBF₃(CF₃), by the perfluoroalkyl radical, Lithium tetrakis(perfluoro carboxylate) borate salts, such as LiB(CF₃COO)₄, LiB(OCOCF₂COO)₂, and LiB(OCOC₂F₄COO)₂, are mentioned.

[0023] These lithium salt may be independent, or two or more kinds may be mixed and it may be used. In the above-mentioned lithium salt, solubility and whenever [ionic dissociation] from the field of a conductivity property LiPF₆, LiBF₄, LiN(CF₃SO₂)₂, LiN(C₂F₅SO₂)₂, LiN(CF₃SO₂)(C₄F₉SO₂), LiPF₃(CF₃)₃, LiPF₃(C₂F₅)₃, LiBF₂(C₂F₅)₂, and LiB(OCOCF₂COO)₂ are desirable, and LiPF₆ or LiBF₄ is still more desirable. When a non-aqueous solvent contains gamma-butyrolactone 60% of the weight or more especially, it is desirable that LiBF₄ is 50% of the weight or more of the whole lithium salt.

[0024] The electrolytic solution is prepared so that the concentration of the lithium salt in the electrolytic solution may become in 1. and 0.5-3 mols /. If concentration is too low, the conductivity of the electrolytic solution will become inadequate, and since conductivity falls by the rise of viscosity or lithium salt becomes easy to deposit at low temperature when concentration is too high, the engine performance of a cell will fall. Occlusion and the ingredient of arbitration which can be emitted can be used for the negative electrode of the cell concerning this invention for a lithium. Carbonaceous ingredient; metallic-oxide ingredient; lithium metals, such as a pyrolysis object of the organic substance in various pyrolysis conditions as an example, an artificial graphite, and a natural graphite; various lithium alloys are mentioned.

[0025] What performed various surface treatment which contains a pitch in these graphites to the artificial graphite and purification natural-graphite list which were manufactured by carrying out elevated-temperature heat treatment of the easy graphite pitch obtained from various raw materials as a carbonaceous ingredient is desirable. That whose d value (distance between layers) of the lattice plane (002nd page) for which it asked by the X diffraction by Gakushin method as a graphite ingredient is 0.335-0.34nm is desirable, and what is 0.335-0.337nm is more desirable. As for ash content, what is 1 or less % of the weight is desirable, its 0.5 or less % of the weight is more desirable, and 0.1 or less % of the weight of its thing is still more desirable. As for the microcrystal size (Lc) for which it asked by the X diffraction by Gakushin method, what is 30nm or more is desirable. Microcrystal size (Lc) has a desirable thing 50nm or more, and what is 100nm or more is more desirable. 1 micrometer - 100 micrometers are desirable, 3 micrometers - 50 micrometers are more desirable, the median size by the method of laser diffracting / being scattered about has 5 micrometers - still more desirable 40 micrometers, and especially its thing that is 7 micrometers - 30 micrometers is desirable. It is 0.5m²/g-25.0m²/g, BET adsorption method specific surface area has desirable g-20.0m²/[of 0.7m²/] g, its g-15.0m²/[of 1.0m²/] g is more desirable, and its thing of g-10.0m²/[of 1.5m²/] g is still more desirable. In the Raman spectrum analysis using Ar-ion-laser light, intensity ratio R=IB/IA of the peak PA of the range of 1580-1620cm⁻¹ (peak intensity IA) and the peak PB (peak intensity IB) of the range of 1350-1370cm⁻¹ is 0-0.5, one or less [25cm⁻¹] comes out [the full width at half maximum of the range of 1580-1620cm⁻¹ / the full width at half maximum of the range of 1 or less / 26cm⁻¹ and 1580-1620cm⁻¹], and a certain thing is desirable.

[0026] Moreover, other negative-electrode material which emits [occlusion and] a lithium can also be mixed and used for a carbonaceous ingredient. as negative-electrode material which emits [occlusion and] lithiums other than a carbonaceous ingredient, metals, such as Ag, Zn, aluminum, Ga, In, Si, germanium, Sn, Pb, P, Sb, Bi, Cu, nickel, Sr, and Ba, and metallic-oxide ingredient; lithium metals of Li, such as oxide of alloy; these metals, are mentioned, among these Sn oxide, Si oxide, aluminum oxide, the lithium alloy of Sn, Si, and aluminum, and a metal lithium are desirable.

[0027] These negative-electrode material may be independent, or two or more kinds may be mixed and used for it. A negative electrode is manufactured by the usual approach. For example, a binder, a thickener, electric conduction material, a solvent, etc. are added to negative-electrode material if needed, and it considers as the shape of a slurry, and it applies to the substrate of a charge collector and a negative electrode is manufactured by drying. Moreover, roll forming of this negative-electrode material is carried out as it is,

and it can consider as a sheet electrode or can also consider as a pellet electrode with compression molding.

[0028] To other ingredients used for the solvent, the electrolytic solution, and the cell which are used at the time of electrode manufacture, if a binder, a thickener, and electric conduction material are stable, they can use the thing of arbitration. Usually, polyvinylidene fluoride, polytetrafluoroethylene, styrene-butadiene rubber, polyisoprene rubber, butadiene rubber, etc. are used as a binder. As a thickener, carboxyl methyl cellulose, methyl cellulose, a hydroxymethyl cellulose, ethyl cellulose, polyvinyl alcohol, oxidation starch, phosphorylation starch, casein, etc. are used.

[0029] As electric conduction material, conductive carbonaceous ingredients, such as metallic material; graphite, such as copper and nickel, and carbon black, are used. Metals, such as copper, nickel, and stainless steel, are mentioned, it is easy to process it into a thin film as the quality of the material of a charge collector in these, and cheap copper foil is desirable. The ingredients including lithium transition-metals multiple oxide ingredients, such as lithium cobalt oxide, a lithium nickel oxide, and a lithium manganic acid ghost, which emit [occlusion and] well-known various lithiums can be used for the positive electrode of a cell. Moreover, a positive electrode can be manufactured according to the negative electrode mentioned above.

[0030] As for the quality of the material of the charge collector for positive electrodes, metals, such as aluminum, titanium, and a tantalum, or the alloy of those is used. In these, aluminum or its alloy is desirable. The thing of arbitration can be used, if it is stable and has sufficient solution retention to the electrolytic solution as a separator which **** the positive electrode and negative electrode of a cell. For example, a porous sheet, a nonwoven fabric, etc. which use polyolefines, such as polyethylene and polypropylene, as a raw material are used.

[0031] Manufacture of the cell concerning this invention can be suitably chosen from the approaches used with the usual nonaqueous electrolyte rechargeable battery. A cell can be made into the configuration of the arbitration used regularly. For example, the coin type which carried out the laminating of the cylinder type, pellet electrode, and separator of the inside-out configuration which combined the cylinder type, pellet electrode, and separator which made the sheet electrode and the separator the shape of a spiral is mentioned.

[0032]

[Example] Although an example and the example of a comparison are given to below and this invention is explained to it still more concretely, this invention is not limited to these examples, unless the summary is exceeded.

(Example 1) The non-aqueous solvent of 95 % of the weight of gamma-butyrolactone and 5 % of the weight of 1-methyl pyrrolidones was prepared, and under desiccation argon atmosphere, this was dissolved so that it might become the rate of one mol/l, and the fully dried lithium fluoride (LiBF₄) was used as the electrolytic solution at it.

[0033] (Example 2) It replaced with 1-methyl pyrrolidone and the electrolytic solution was prepared like the example 1 except having used 3-methyl-2-oxazolidone.

(Example 3) It replaced with 1-methyl pyrrolidone and the electrolytic solution was prepared like the example 1 except having used 1,3-dimethyl-2-imidazolidinone.

[0034] (Example 4) It replaced with 1-methyl pyrrolidone and the electrolytic solution was prepared like the example 1 except having used 1-vinyl pyrrolidone.

(Example 5) Prepared 90 % of the weight of gamma-butyrolactone, 5 % of the weight of 1-vinyl pyrrolidone, and the non-aqueous solvent of 5 % of the weight of vinylene carbonate, and it was made to dissolve so that it may become this in 1. and one mol /about LiBF₄, and considered as the electrolytic solution.

[0035] (Example 6) The non-aqueous solvent of 19 % of the weight of gamma-butyrolactone, 19 % of the weight of ethylene carbonate, 60 % of the weight of ethyl methyl carbonate, and 2 % of the weight of 1-methyl pyrrolidones was prepared, LiBF₄ fully dried under desiccation argon atmosphere was dissolved so that it might become [1.] in one mol /, and it considered as the electrolytic solution.

[0036] (Example 7) 18 % of the weight of gamma-butyrolactone, 18 % of the weight of ethylene carbonate, 60 % of the weight of ethyl methyl carbonate, 2 % of the weight of 1-methyl pyrrolidones, and the non-aqueous solvent of 2 % of the weight of vinylene carbonate were prepared, and under desiccation argon atmosphere, this was dissolved so that it might become [1.] in one mol /, and fully dried LiBF₄ was used as the electrolytic solution at it.

[0037] (Example 8) 18 % of the weight of gamma-butyrolactone, 18 % of the weight of ethylene carbonate, 60 % of the weight of ethyl methyl carbonate, 2 % of the weight of 1-methyl pyrrolidones, and the non-aqueous solvent of 2 % of the weight of vinylene carbonate were prepared, and under desiccation argon atmosphere, this was dissolved so that it might become [1.] in one mol /, and the fully dried 6 phosphorus-fluoride acid lithium (LiPF₆) was used as the electrolytic solution at it.

[0038] (Example 9) Under desiccation argon atmosphere, further, fully dried LiBF₄ was dissolved so that it might become [1.] in 0.5 mols /, this was dissolved so that it might become [1.] in 0.5 mols /, 18 % of the weight of gamma-butyrolactone, 18 % of the weight of ethylene carbonate, 60 % of the weight of ethyl methyl carbonate, 2 % of the weight of 1-methyl pyrrolidones, and the non-aqueous solvent of 2 % of the weight of vinylene carbonate were prepared, and LiPF₆ fully dried under desiccation argon atmosphere was used as the electrolytic solution at it.

[0039] (Example 1 of a comparison) LiBF₄ was dissolved in gamma-butyrolactone so that it might become in 1. and one mol /, and the electrolytic solution was prepared.

(Example 2 of a comparison) 95 % of the weight of gamma-butyrolactone and the non-aqueous solvent of 5 % of the weight of vinylene carbonate were prepared, LiBF₄ was dissolved so that it might become in 1. and one mol /, and it considered as the electrolytic solution.

[0040] (Example 3 of a comparison) LiPF₆ was dissolved in propylene carbonate so that it might become in 1. and one mol /, and the electrolytic solution was prepared.

(Example 4 of a comparison) The non-aqueous solvent of 95 % of the weight of propylene carbonate and 5 % of the weight of vinylene carbonate was prepared, LiPF₆ was dissolved so that it might become in 1. and one mol /, and it considered as the electrolytic solution.

(Production of a positive electrode) 6 % of the weight of carbon black and polyvinylidene fluoride KF-1000(Kureha chemistry company make, trade name) 9 % of the weight were added to the LiCoO₂ 85% of the weight as positive active material, and it mixed, and distributed by 1-methyl pyrrolidone, and what was made into the shape of a slurry was applied to homogeneity on aluminium foil with a thickness of 20 micrometers which is a positive-electrode charge collector, and after desiccation, it pierced to discoid with a

diameter of 12.5mm, and considered as the positive electrode.
d value of the lattice plane (002nd page) in an X diffraction 0.336nm, (Production of a negative electrode) 100nm or more (264nm) and ash content 0.04 % of the weight, [crystallite size (Lc)] 17 micrometers and BET adsorption method specific surface area 8.9m²/g, [the median size by the method of laser diffracting / being scattered about] In the Raman spectrum analysis using Ar-ion-laser light intensity ratio R=IB/IA of the peak PA of the range of 1580-1620cm⁻¹ (peak intensity 1A) and the peak PB (peak intensity IB) of the range of 1350-1370cm⁻¹ 0.15 and artificial-graphite powder KS-44 whose full width at half maximum of the range of 1580-1620cm⁻¹ is 22.2cm⁻¹ (the TIMCAL, LTD. make, trade name) The styrene butadiene rubber (SBR) made to distribute to 94% of the weight with distilled water was added so that it might become 6 % of the weight by solid content. This was mixed by the De Dis parser and it considered as the shape of a slurry, and it applied to homogeneity on copper foil with a thickness of 18 micrometers which is a negative-electrode charge collector, and after desiccation, it pierced to discoid with a diameter of 12.5mm, and considered as the negative electrode.

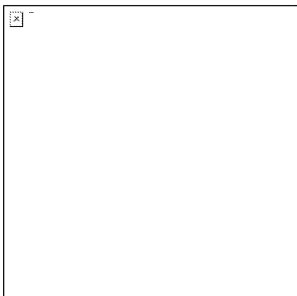
(Production of a coin mold cel) The cell was produced using a positive electrode, an above-mentioned negative electrode, and the above-mentioned electrolytic solution. The positive electrode was held in the can made from stainless steel which serves as a positive-electrode conductor, and the negative electrode was laid through the separator made from polyethylene which infiltrated the electrolytic solution on it. the obturation plate which serves both as this can and a negative-electrode conductor -- the gasket for an insulation -- minding -- it sealed in total and the coin mold cel was produced.

(Evaluation of a coin mold cel) In 25 degrees C, the value which performed the charge and discharge test by charge termination electrical-potential-difference 4.2V and discharge-final-voltage 2.5V or 0.5mA constant current, and broke the discharge capacity of a two-cycle eye by charge capacity of a two-cycle eye was made into two-cycle eye charge-and-discharge effectiveness.

[0041] Moreover, after charging 5 cycle eye on the same conditions, in the state of charge, at 85 degrees C, it saved for 72 hours and the afterdischarge was carried out. The value which broke by charge capacity of a four-cycle eye the value which charged 6 cycle eye on the same conditions by 25 degrees C again was made into the capacity maintenance factor. The rechargeable battery manufactured by the approach mentioned above was evaluated using the electrolytic solution of examples 1-9 and the examples 1-4 of a comparison. A result is shown in Table 1.

[0042]

[Table 1]



[0043]

[Effect of the Invention] The safe nonaqueous electrolyte rechargeable battery in which the cell property which was high, was excellent in the capacity maintenance property, and was excellent in the large temperature requirement with this invention is shown can be offered. [of charge-and-discharge effectiveness]

[Translation done.]
